

526 Rec'd PCT/PTO 04 OCT 2000  
09/647743

Atty. Docket #: 1998/G-004

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

**INTERNATIONAL APPL. NO.:** PCT/EP99/02280 :

**INTERNATIONAL FILING DATE:** -04/01/99- :

**APPLICANT:** STEFAN DISCH ET AL :

**SERIAL NO:** : **ART UNIT:**

**FILED:** -HEREWITH- : **EXAMINER:**

**FOR:** "POLYOXYMETHYLENE MOLDING COMPOSITION WITH IMPROVED STABILITY DURING PROCESSING AND REDUCED EMISSION TENDENCY" :

Assistant Commissioner for Patents  
Box PCT  
Washington, D.C. 20231

"Express Mail" No.: EK219527559

Date: - OCTOBER 04, 2000 -

I hereby certify that this paper, along with any other paper or fee referred to in this paper as being transmitted herewith, is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10, postage prepaid, on the date indicated above, addressed to the Asst. Comm. for Patents, Washington, D.C. 20231

- Kay P. Ruark -  
(Typed or printed name of mailing paper or fee)

  
(Signature of person mailing paper)

**TRANSMITTAL OF APPLICATION PAPERS  
TO U.S. DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. §371  
(37 CFR 1.494 OR 1.495)**

This Transmittal Letter is based upon PTO Form 1390 (as revised in May, 1993).

The above-identified applicant(s) (jointly with their assignee) have filed an International Application under the P.C.T. and hereby submit(s) to the United States Designated/Elected Office (DO/EO/US) the following items and other information.

1.  This is a **FIRST** submission of items concerning a filing under 35 U.S.C. §371.
2.  This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. §371.
3.  This is an express request to begin national examination procedures (35 U.S.C. §371[f]) at any time rather than delay.
4.  A proper Demand for International Preliminary Examination (IPE) was made to the appropriate Authority (IPEA) within the time period required.
  
5.  A copy of the International Application as filed (35 U.S.C. §371[c][2]) --
  - a.  is transmitted herewith (required when not transmitted by International Bureau). See WIPO Publication WO 99/52959.
  - b.  has been transmitted by the International Bureau.
  - c.  is not required, as the application was filed in the United States Receiving Office (RO/US).
6.  A (verified) translation of the International Application into the English language is enclosed.
7.  Amendments to the (specification and) claims of the International Application under PCT Article 19 (35 U.S.C. 371[c][3])
  - a.  are transmitted herewith (required if not transmitted by the International Bureau).
  - b.  have been transmitted by the International Bureau.
  - c.  have not been made; however, the time limit for making such amendments has NOT expired.
  - d.  have not been made and will not be made.
  - e.  will be submitted with the appropriate surcharge.
8.  A translation of the amendments to the claims (and/or the specification) under PCT Article 19 (35 U.S.C. §371[c][3]) is enclosed or will be submitted with the appropriate surcharge.

9.  An oath or declaration/power of attorney of the inventor(s) (35 U.S.C. §371[c][4]) is enclosed.  
 and is attached to the translation of (or a copy of) the International Application.  
 and is attached to the substitute specification.

10.  A translation of at least the Annexes to the IPE Report under PCT Article 36 (35 U.S.C. §371[c][5]) is enclosed.

**Items 11. to 16. below concern other document(s) or information included:**

11.  An Information Disclosure Statement under 37 CFR 1.97 and 1.98 is enclosed.

12.  An Assignment for recording and a separate cover sheet in compliance with 37 CFR 3.28 and 3.31 are enclosed.

13.  A FIRST preliminary amendment is enclosed.  
A SECOND or SUBSEQUENT preliminary amendment is enclosed.

14.  A substitute specification (including claims, abstract, drawing) is enclosed.

15.  A change of power of attorney and/or address letter is enclosed.

16.  Other items of information:

This application is being filed pursuant to 37 CFR 1.494(c) or 1.495(c), and any missing parts will be filed before expiration of--

22 months from the priority date under 37 CFR 1.494(c), or

32 months from the priority date under 37 CFR 1.495(c).

The undersigned attorney is authorized by the International applicant and by the inventors to enter the National Phase pursuant to 37 CFR 1.494(c) or 1.495(c).

The following additional information relates to the International Application:

09/647743  
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International Application No. PCT/EP99/02280

1998/G-004

- Receiving Office: EPO
- IPEA (if filing under 37 CFR 1.495): EPO
- Priority Claim(s) (35 USC §§ 119, 365):  
German Appln. 198 15 663.4 dated April 08, 1998.
- A copy of the International Search Report is
  - enclosed.
  - attached to the copy of the International Application.
- A copy of the Receiving Office Request Form is enclosed.
- Form PCT/IB/308 (1) sheet

The fee calculation is set forth on the next page of this Transmittal Letter.

## FEE CALCULATION SHEET

A check in payment of the filing fee, calculated as follows, is attached (37 CFR 1.492).

Basic Fee..... \$ 860.00

Total Number of claims in  
excess of (20) times \$18..... -0-

Number of independent claims  
in excess of (3) times \$80..... -0-

Fee for multiple dependent  
claims \$270..... -0-

TOTAL FILING FEE... \$ 860.00

Kindly send us the official filing receipt.

The Commissioner is hereby authorized to charge any additional fees which may be required or to credit any overpayment to Deposit Account No. 03-2775. This is a "general authorization" under 37 CFR 1.25(b), except that no automatic debit of the issue upon allowance is authorized. An additional copy of this page is attached.

Respectfully submitted,

By Richard M. Beck

Richard M. Beck

Reg. No. 22,580

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RMB/kpr (5587\*264)

Enclosures

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EMISSION TENDENCY"

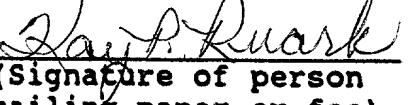
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.....  
Assistant Commissioner  
for Patents  
Washington, D.C. 20231

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- Kay P. Ruark -  
(Typed or printed name) of  
person mailing paper or fee)

  
(Signature of person  
mailing paper or fee)

PRELIMINARY AMENDMENT

Sir:

Prior to any action on the merits of the accompanying  
new patent application, kindly amend the application as  
follows:

In the Claims:

Claim 3, line 1, delete "or 2" ;

Claim 4, line 1, change "any of claims 1 to 3" to read  
-- claim 1 -- ;

**Claim 5**, lines 1 and 2, change "any one of claims 1 to 4" to read -- claim 1 -- ;

**Claim 6**, lines 1 and 2, change "any one of claims 1 to 4" to read -- claim 1 -- ;

**Claim 7**, line 1, delete "or 6" ;

**Claim 8**, line 1, delete "or 6" ;

**Claim 9**, line 1, delete "or 6" ;

**Claim 10**, line 1, delete "or 6" .

**R E M A R K S**

**Claims 3-10** have been amended to refer to only one preceding claim. Each of the dependent claims, as amended, now depends on only one preceding claim. Therefore no additional fee is required for multiple dependency.

Prompt, favorable action is solicited.

Respectfully submitted,

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Description

Polyoxymethylene molding composition with improved stability during processing and reduced emission tendency

5

The present invention relates to a polyoxymethylene molding composition which has particularly high stability, and to its use for producing low-emission moldings. These materials are particularly suitable for producing colored moldings with low emissions.

10

Since they were introduced to the market about 30 years ago, polyoxymethylenes (POMs) have become established as extremely useful materials for a variety of technical applications. POM is particularly widely used as a material in designs for automotive construction or for the 15 electrical industry. Examples can be found in the technical service brochures of POM producers.

20

POM copolymers and their preparation are by now well known (Sabel et al., in Becker/Braun Eds., Kunststoff-Handbuch, Vol. 3/1). For example, it is now well known that trioxane can be copolymerized with cyclic ethers using cationic initiators. The usual cationic initiators used are Lewis acids, such as  $\text{BF}_3$ , strong protonic acids, such as  $\text{HClO}_4$ , heteropolyacids or perfluoroalkanesulfonic acids. The comonomer used is usually ethylene oxide or the cyclic formal of ethylene glycol, butanediol or diethylene glycol.

25

In principle the comonomer content in the POM copolymer can be varied very widely. For example, JP07286024 gives a range of from 0.03 to 10 mol% of comonomer units in the polymer for copolymers of oxymethylene and C<sub>2</sub>-C<sub>4</sub>-oxyalkylenes modified with long-chain aliphatic 30 end groups. JP07124996 gives a general description of a POM copolymer in which the proportion of comonomer in the polymer may be from 3 to 30% by weight.

35

The comonomer content generally given for the preparation of POM copolymers is from 3 to 4% by weight, and this data is to be understood as the proportion by weight of the comonomer in the monomer mixture (examples: JP07286023; JP06049155; JP04108819).

To obtain products of greater stability, POM molding compositions are treated for a prolonged period with aqueous ammonia solution (JP54107972) or heated in aqueous suspension under pressure at from 100 to 200°C (NL-A6812966).

5

Products made from POM copolymers have long been produced commercially and used for engineering components. A certain level of mechanical properties, such as stiffness, hardness and toughness, is required here from POM molding compositions, and it is only this which 10 allows the use of these materials for engineering components such as gear wheels, levers and many others. The yield stress values published in the brochures of POM copolymer manufacturers are from 60 to 70 N/mm<sup>2</sup>. The values found there for the modulus of elasticity of unmodified copolymers are from 2400 to 3100 N/mm<sup>2</sup>. The values found for notched 15 impact strength at 23°C are from 4 to 12 mJ/mm<sup>2</sup>.

Because POM molding compositions have these advantageous properties there is the requirement to give these materials access to more application sectors. A demand increasingly met with, alongside maintenance of the 20 mechanical property profile, is that there should only be very little emission of residual monomers or of other volatile constituents from moldings. The automotive industry, one of the most important markets for products made from POM, has developed specific analysis methods for this purpose (VDA Empfehlung Nr. 275 [German Automotive Industry Recommendation No. 25 275], documented by Kraftfahrwesen e.V., July 1994). A low proportion of residual monomers and of other volatile constituents is also important with respect to the coloration of POM, since POM is particularly difficult to color (cf. Damm W., Herrmann E., in Gächter, Müller; 3rd Edition, p. 730).

30 Although the commercially produced POM products currently obtainable have the known advantageous mechanical properties, the moldings produced from these have excessive emission of 30 mg/kg or more of formaldehyde. Attempts are made to achieve a lower emission level by complicated post-treatment of the moldings, e.g. by intensive annealing. 35 However, the post-treatment gives rise to additional costs.

The POM stabilization systems described in a variety of patents, generally using certain formaldehyde scavengers, have hitherto also been unable to

overcome the defect of high emissions. Low-emission moldings cannot be produced reliably from the POM molding compositions which have hitherto been customary.

5 There was therefore a need to develop POM molding compositions which firstly have a substantially reduced emission tendency and secondly have the level of mechanical properties which is known and required by the industry.

10 The object is achieved by linear POM copolymers which essentially have oxymethylene units and oxyethylene units as structural units in the polymer chain, where the proportion of oxyethylene units in the structural units of the polymer chain is from 1.5 to 2.5 mol%, preferably from 1.85 to 2.25 mol%.

15 Surprisingly, the novel molding compositions specifically have firstly substantially improved stability, so that the residual emission levels of moldings produced from these are extremely low. For example, formaldehyde emission, measured on sheets of wall thickness 1 mm after 20 24 hours of storage, in accordance with VDA 275, is generally less than 15 mg/kg, preferably less than 10 mg/kg.

25 Secondly, the mechanical properties of the novel molding compositions meet the customary requirements placed upon commercially available POM products, and they can therefore be utilized without restriction for the application sectors and processing techniques customary for POM.

30 The novel molding compositions are composed of linear POM copolymers which essentially have only oxyethylene units as co-component in the polymer chain alongside oxymethylene units. In principle, the copolymer may also contain a small proportion of longer-chain units, e.g. C<sub>3</sub>- or C<sub>4</sub>-oxyalkylene units. However, these impair mechanical properties at a constant low emission tendency.

35 In the novel molding compositions, the proportion of oxyethylene units in the structural units of the polymer chain is generally 2.0 ± 0.5 mol%, i.e. from 1.5 to 2.5 mol%. Their proportion is preferably from 1.85 to 2.25 mol%. The proportion of longer-chain oxyalkylene units should

generally not exceed 0.6 mol%, preferably 0.3 mol%, of the structural units of the polymer chain. The total of oxyethylene units and longer-chain oxyalkylene units in the structural units of the polymer chain is also generally from 1.5 to 2.5 mol%, preferably from 1.85 to 2.25 mol%.

5

Although POM copolymers whose proportion of comonomer units is higher than that of the novel products likewise have the required low emission values, their substantially lower stiffness and strength values mean that they cannot be used in the application sectors known for POM.

10

The novel POM copolymers may be prepared by well known preparation processes. An example of a possible process is the copolymerization of trioxane with from 4 to 6% by weight, preferably from 4.5 to 5.5% by weight, of dioxolane in the presence of generally customary amounts of 15  $\text{BF}_3$  and methylal, where the amount of dioxolane is based on the total of dioxolane and trioxane. The proportion of the comonomer in the monomer mixture is correspondingly from 4.8 to 7.2 mol%, preferably from 5.4 to 6.6 mol%.

20

The customary stabilizers and auxiliaries, such as antioxidants, mold-release aids, acid scavengers, nitrogen-containing costabilizers and nucleating agents may be added either individually or as a mixture to the novel POM copolymers, if desired together with colorants. The addition of stabilizers may, however, also be reduced or dispensed with, since the 25 stability of the novel copolymers is in any case high.

30

The extremely low emission values of the novel POM copolymer molding compositions mean that they can particularly advantageously be used directly for producing low-emission moldings. No post-treatment of the moldings by annealing is now required, and their overall production is therefore more cost-effective.

35

Particular application sectors for the novel molding compositions are internal fittings and claddings of means of transport, such as automobiles, aircraft, railroad cars, etc., household products, toys, in particular children's toys, items for babies, and also devices and components for electrical engineering and electronics. The novel molding composition is particularly suitable for producing colored moldings and for producing low-emission

apparatuses and instruments, or parts of these, for medical applications, for example inhalers.

### Examples

5

In the examples which follow the properties of materials were determined by the following methods:

10 Melt index in accordance with ISO 1133 at 190°C and with a weight of 2.16 kg applied;

15 modulus of elasticity in accordance with ISO 527;

20 yield stress in accordance with ISO 527;

25 notched impact strength in accordance with ISO 179;

30 formaldehyde emission: sheets of wall thickness 1 mm were manufactured from the POM copolymer molding compositions. The formaldehyde emission from the sheets was determined in accordance with VDA 275 after 24 hours in storage.

35 The results of the materials testing for the examples below are given in Table 1.

#### Example 1

3400 g of trioxane were copolymerized with 190 g of dioxolane in the presence of 35 ppm of  $\text{BF}_3$  and 1200 ppm of methylal. After removal of 25 unconverted monomers, initiator residues and unstable end groups were removed by dissolving and heating the copolymer in a methanol/water/trioxane mixture at 180°C at superatmospheric pressure. The proportion of oxyethylene units in the resultant copolymer was 2.05 mol%. This polymer was melted in a kneader and mixed with 30 antioxidant, acid scavenger and additives in the same proportions as in the comparative polymer. Pellets were produced from the mixture, and these were injection molded to give the test specimens for determining the modulus of elasticity, yield stress and notched impact strength, and to give the sheets for determining formaldehyde emission. The melt index was 35 likewise determined on the mixture.

**Comparative Example 1**

3400 g of trioxane were copolymerized with 275 g of dioxolane in the presence of 35 ppm of  $\text{BF}_3$  and 1200 ppm of methylal. After removing unconverted monomers, initiator residues and unstable end groups were

5 removed as in Example 1. The proportion of oxyethylene units in the resultant copolymer was 3.1 mol%. The copolymer was melted as in Example 1, mixed with antioxidant, acid scavenger and additives, pelletized and molded to give test specimens.

**10 Comparative Example 2**

Commercially available POM copolymer (Hostaform<sup>®</sup>) with a melt index comparable to that of Example 1 at 28 g/10 min, was used as in Example 1 to manufacture sheets of wall thickness 1 mm, of which the formaldehyde emission was determined.

**15 Example 2**

3400 g of trioxane were copolymerized with 180 g of dioxolane in the presence of 35 ppm of  $\text{BF}_3$  and 800 ppm of methylal. After removing unconverted monomers, initiator residues and unstable end groups were

20 removed as in Example 1. The proportion of oxyethylene units in the resultant copolymer was 1.92 mol%. The copolymer was melted as in Example 1, mixed with antioxidant, acid scavenger and additives, pelletized and molded to give test specimens.

**25 Comparative Example 3**

Commercially available POM copolymer (Hostaform<sup>®</sup>) with a melt index comparable to that of Example 2 at 13 g/10 min, was used as in Example 1 to manufacture sheets of wall thickness 1 mm, of which the formaldehyde emission was determined.

**30 Example 3**

3400 g of trioxane were copolymerized with 200 g of dioxolane in the presence of 35 ppm of  $\text{BF}_3$  and 600 ppm of methylal. After removing unconverted monomers, initiator residues and unstable end groups were

35 removed as in Example 1. The proportion of oxyethylene units in the resultant copolymer was 2.13 mol%. The copolymer was melted as in Example 1, mixed with antioxidant, acid scavenger and additives, pelletized and molded to give test specimens.

### Comparative Example 4

3400 g of trioxane were copolymerized with 280 g of dioxolane in the presence of 35 ppm of  $\text{BF}_3$  and 600 ppm of methylal. After removing unconverted monomers, initiator residues and unstable end groups were removed as in Example 1. The proportion of oxyethylene units in the resultant copolymer was 3.22 mol%. The copolymer was melted as in Example 1, mixed with antioxidant, acid scavenger and additives, pelletized and molded to give test specimens.

10

### Comparative Example 5

Commercially available POM copolymer (Hostaform®) with a melt index comparable to that of Example 3 at 9 g/10 min, was used as in Example 1 to manufacture sheets of wall thickness 1 mm, of which the formaldehyde emission was determined.

#### Example 4

3400 g of trioxane were copolymerized with 200 g of dioxolane in the presence of 35 ppm of  $\text{BF}_3$  and 1600 ppm of methylal. After removing unconverted monomers, initiator residues and unstable end groups were removed as in Example 1. The proportion of oxyethylene units in the resultant copolymer was 2.08 mol%. The copolymer was melted as in Example 1, mixed with antioxidant, acid scavenger and additives, pelletized and molded to give test specimens.

25

### Comparative Example 6

Commercially available POM copolymer (Hostaform<sup>®</sup>) with a melt index comparable to that of Example 4 at 50 g/10 min, was used as in Example 1 to manufacture sheets of wall thickness 1 mm, of which the formaldehyde emission was determined.

Table 1		Oxyethylene units mol%	Melt index g/10 min	Modulus of elasticity N/mm <sup>2</sup>	Yield stress N/mm <sup>2</sup>	Notched impact strength mJ/mm <sup>2</sup>	Formaldehyde emission mg/kg
Example 1	2.05	27	2650	61	7	7	7
Comparative Example 1	3.10	28	2100	52	9	5	5
Comparative Example 2	1.31	28				35	
Example 2	1.92	13	2680	62	6.5	8.2	8.2
Comparative Example 3	1.43	13				30	
Example 3	2.13	9	2580	60	7.5	6.5	6.5
Comparative Example 4	3.22	9	2050	50	9	4	4
Comparative Example 5	1.34	9				33	
Example 4	2.08	50	2580	60	7.5	6.5	6.5
Comparative Example 6	1.32	50				33	

## Claims

1. A molding composition made from linear polyoxymethylene copolymers which essentially have oxymethylene units and oxyethylene units as structural units in the polymer chain, where the proportion of oxyethylene units in the structural units of the polymer chain is from 1.5 to 2.5 mol%, preferably from 1.85 to 2.25 mol%.
2. A molding composition as claimed in claim 1, which has a formaldehyde emission, measured on sheets of wall thickness 1 mm after 24 hours in storage, in accordance with VDA 275, of less than 15 mg/kg, preferably less than 10 mg/kg.
3. A molding composition as claimed in claim 1 or 2, which has a modulus of elasticity in accordance with ISO 527 of from 2400 to 3100 N/mm<sup>2</sup>, a yield stress in accordance with ISO 527 of from 60 to 70 N/mm<sup>2</sup> and a notched impact strength in accordance with ISO 179 at 23°C of from 4 to 12 mJ/mm<sup>2</sup>.
4. A molding composition as claimed in any of claims 1 to 3, which comprises antioxidants, acid scavengers, stabilizers and colorants.
5. The use of a molding composition as claimed in any one of claims 1 to 4 for producing moldings which have a formaldehyde emission of less than 15 mg/kg, preferably less than 10 mg/kg.
6. The use of a molding composition as claimed in any one of claims 1 to 4 for producing colored moldings.
7. The use as claimed in claim 5 or 6 for low-emission moldings for interior fittings or interior cladding in means of transport, such as automobiles, aircraft and rail road cars.
8. The use as claimed in claim 5 or 6, for household products, recreational items, in particular children's toys, and items for babies.
9. The use as claimed in claim 5 or 6 for devices and components for electrical engineering and electronics.

10. The use as claimed in claim 5 or 6 for apparatuses and instruments for medical applications.

Abstract

Molding compositions made from linear polyoxymethylene copolymers which essentially have oxymethylene units and oxyethylene units as structural units in the polymer chain, where the proportion of oxyethylene units in the structural units of the polymer chain is from 1.5 to 2.5 mol%. These molding compositions, and also uncolored or colored moldings produced from these have high stability and extremely low emission of formaldehyde and of residual monomers. At the same time they have a high level of mechanical properties, and they can therefore be utilized without restriction for the customary application sectors and processing methods.

**COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY**

As below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below, I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

**Polyoxymethylene molding composition with improved stability during processing and reduced emission tendency**

the specification of which

- is attached hereto
- was filed on as

and including all the amendments through the date hereof.

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

**Prior Foreign Application(s) for which Priority is Claimed:**

Federal Republic of Germany, 19815663.4 of April 8, 1998

And I hereby appoint

Rudolf E. Hutz, Reg.No. 22,397; Harold Pezzner, Reg.No. 22,112; Richard M. Beck, Reg.No. 22,580; Paul E. Crawford, Reg.No. 24,397; Thomas M Meshbesher, Reg.No. 25,083; Robert G. McMorrow, Jr., Reg.No. 30,962; Patricia Smink Rogowski, Reg.No. 33,791; Ashley I. Pezzner, Reg.No. 35,646; William E. McShane, Reg. 32,707; James T. Moore, Reg. No. 35,619; Mary W. Bourke, Reg.No. 30,982; Gerard M. O'Rourke, Reg.No. 39,794; Christine M. Hansen, Reg.No. 40,634; Allan N. Kutzenco, Reg. No. 38,945; James M. Olsen, Reg.No. 40,408; Francis DiGiovanni, Reg.No. 37,310; Frank Z. Yang, Reg. No. 35,417; Eric J. Vain, Reg.No. 42,517; Daniel C. Mulveny, Reg.No. P-45,897; and Elliot C. Mendelson, Reg.No. 42,878

20  
all of CONNOLLY AND HUTZ, P.O.Box 2207, Wilmington, Delaware 19899-2007, my attorneys with full power of substitution, to prosecute this application, and transact all business in the Patent and Trademark Office connected therewith and I hereby request that all correspondence in this application be directed to:

**CONNOLLY AND HUTZ**  
**P.O.Box 2207**  
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**Telephone (302) 658-9141**

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

INVENTOR(S) / Residence

1 - ∞ 1) Dr. Stefan Disch Hainerweg 24, 60599 Frankfurt, Germany DEX

Signature: Stefan Disch Date: 11/8/00

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Signature: \_\_\_\_\_ Date: \_\_\_\_\_

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Signature: Karl-Friedrich Mück Date: 11/08/2000

4) Gerhard Reuschel Sulzbacher Straße 11, 65835 Liederbach, Germany DEX

Signature: Gerhard Reuschel Date: 16/8/2000

5) Lothar Reißmann, Am Pfingstbrunnen 22, 65719 Hofheim, Germany

Signature: \_\_\_\_\_ Date: \_\_\_\_\_

The inventors are citizens of Germany.

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Germany

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

INVENTOR(S) / Residence

1) Dr. Stefan Disch Hainerweg 24, 60599 Frankfurt, Germany

Signature: \_\_\_\_\_ Date: \_\_\_\_\_

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Signature: Peter Eckhardt Date: 9/1/2000

3) Dr. Karl-Friedrich Mück Schnitterweg 7, D-65207 Wiesbaden, Germany

Signature: \_\_\_\_\_ Date: \_\_\_\_\_

4) Gerhard Reuschel Sulzbacher Straße 11, 65835 Liederbach, Germany

Signature: \_\_\_\_\_ Date: \_\_\_\_\_

5) Lothar Reißmann, Am Pfingstbrunnen 22, 65719 Hofheim, Germany

Signature: \_\_\_\_\_ Date: \_\_\_\_\_

The inventors are citizens of Germany.

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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